COINCIDENCE-MÖSSBAUER SPECTROSCOPY*

Dennis W. Hamill† and Gilbert R. Hoy‡
Physics Department, Boston University, Boston, Massachusetts
(Received 19 July 1968)

We show that in analyzing coincidence-Mössbauer data it is necessary to include the finite resolution time determined by the electronics. By so doing, we are able to fit spectra in which only "time filtering" effects are present. Results are also presented showing the enhanced resolution capability of this technique.

The coincidence-Mössbauer technique can be summarized for iron as follows: Co^{57} undergoes electron capture to form a second excited state of Fe^{57} which decays by emission of a 122-keV gamma ray to the 14.4-keV Mössbauer level. The detection of the 122-keV gamma ray signals the occupation of the intermediate state, thus allowing the experimenter to use the Mössbauer effect selecting only those 14-keV gamma rays that are emitted during some preset time interval. Since not all 14-keV gamma rays are recorded, "time filtering" effects result in line shapes that are non-Lorentzian. The nature of this "time filtering" has been studied previously[1-3] and is understood on both a classical and quantum mechanical basis.[2,4] However, a quantitative fit of theory and experiment in those solid-state systems in which only time-filtering effects should dominate has not previously been accomplished.[1-3,5]

The expression for transmission $I(T)$ through a resonant absorber as a function of time can be written conveniently in two forms depending on the time region of interest.[2] The form

$$I(T)=e^{-T}\left|\sum_{n=0}^{\infty}\left(\frac{4i\Delta\omega}{\beta\Gamma}\right)^{n}\left(\frac{\beta T}{4}\right)^{\frac{1}{2}n}J_{n}(\beta^{\frac{1}{2}}T^{\frac{1}{2}})\right|^{2} \tag{1}$$

converges rapidly for $(2\Delta\omega\beta^{1/2}T^{1/2})/(\beta\Gamma)<1$, while

$$I(T)=e^{-T}\left|-\exp\left[i\left(\frac{\Delta\omega T}{\Gamma}+\frac{\Gamma\beta}{4\Delta\omega}\right)\right]+\sum_{n=1}^{\infty}\left(\frac{i\beta\Gamma}{4\Delta\omega}\right)^{n}\left(\frac{\beta T}{4}\right)^{-\frac{1}{2}n}J_{n}(\beta^{\frac{1}{2}}T^{\frac{1}{2}})\right|^{2} \tag{2}$$

is appropriate for $(2\Delta\omega\beta^{1/2}T^{1/2})/(\beta\Gamma)>1$. Here T is time in nuclear lifetime units, Γ is the natural linewidth in mm/sec, $\Delta\omega$ is the frequency difference between the emission and absorption lines, J_n's are integer Bessel functions, and $\beta=n\sigma_0 f$ is the absorber thickness (n = the number of Fe^{57} atoms/cm^2, σ_0 = the resonance cross section, and f = the recoilless fraction in the absorber). Previous efforts to fit experimental data have used this expression evaluated at a particular relevant instant in time. The inability to fit the data using this approach led Neuwirth[5] to investigate the consequences of postulating a nonhomogenous absorber and hence resulted in averaging over β according to various assumed distributions. However, the results were not totally satisfactory. We will show that this lack of success is due to neglect of the fact that experimentally one looks at the transmission over an interval in time determined by the electronics and adjustable within certain limits. Our calculations differ from those done previously in that they involve a numerical integration of the exact expressions given above over the experimental time window.

Figure 1 illustrates what happens for a particular time T as the time window is opened. The precise nature of the result depends on the time T. Generally, the most obvious effect of the integration is the rapid damping of absorption oscillations due to interfering contributions from different times in the window. Moreover, as shown in Fig. 1, it is possible that quite far from the central peak the contributions reinforce each other producing a recurrence.

In order to check our analytical procedures, the exact expression for the time-dependent transmission was numerically integrated over all times (in actuality from 0 to 1000 nsec). These results were then compared with those of Margulies and Ehrman[6] who derived an expression for linewidth as a function of their effective thickness parameter which is identical to the parameter β in Eqs. (1) and (2). The agreement was excellent and the results of our calculations were

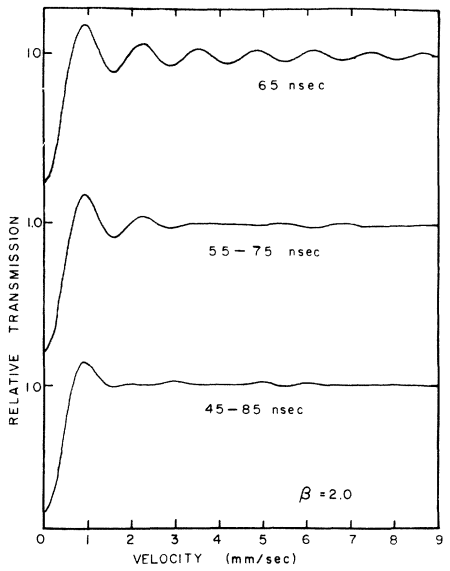

FIG. 1. Calculated coincidence-Mössbauer spectra as a function of the time-window width.

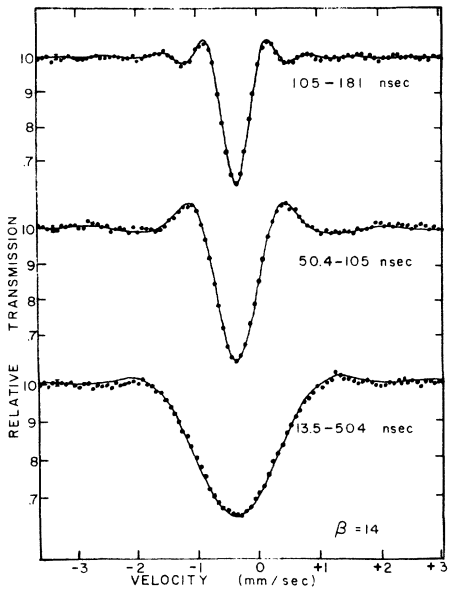

FIG. 2. Experimental and theoretical results for the single-line source, Co^{57} in copper.

then extended to thicker absorbers. This consistent result gives a check on the value of β and hence removes it as a free parameter in the coincidence-Mössbauer analysis.

Our experimental technique was similar to that of other experimenters mentioned above. The detected 122-keV gamma ray was used to start a time-to-pulse-height converter which was stopped by the detection of the 14-keV gamma ray. The output pulses were fed into single-channel analyzers each of which selected a range of pulse heights corresponding to a particular time window. The outputs of these single-channel analyzers were used to route into different quadrants of a Nuclear Data 512-channel analyzer. The particular channel in a quadrant was determined by the source velocity at the time of arrival of the 14-keV gamma ray. Spectra for three different time windows were taken at once, with the fourth quadrant being used to store a regular Mössbauer spectrum. This regular spectrum was used in the analysis to normalize for background shape and chance coincidences. The chance coincidence rate and the time window settings were determined by taking decay curves of the 14-keV nuclear transition.

Figure 2 shows our experimental and theoretical results for a single-line source (Co^{57} in Cu) and absorber $[Na_4Fe(CN)_6]$. The solid curves were obtained by integrating the time-dependent transmission over the predetermined time window as given in the figure. The value of β was calculated from the known thickness of the absorber (1.0 mg/cm^2 of Fe^{57}). There are no free parameters. The agreement between theory and experiment establishes the necessity of adopting our procedure when analyzing even this simplest type of coincidence-Mössbauer spectrum.

In a search for time-dependent solid-state effects, one must deal with multiline spectra. It should be clear that extreme care may be needed in order to extract accurate line positions and intensities particularly in view of the previously noted recurrences. We have mentioned this earlier in connection with the study of "after effects."[7,8]

We will now show that even when one is interested in the physics of the absorber, it may be advisable to use coincidence-Mössbauer techniques. This technique enhances the experimental percentage effect and permits increased resolution. The former effect is simply a result of recording only coincidence pulses and thus eliminating much of the noise which contributes background in a regular Mössbauer spectrum taken using scintillation detectors. Improvement in the resolution is obtained by taking spectra counting only those gamma rays coming from nuclei that have lived longer than one lifetime and thus have a better defined energy. Previous attempts[9] to demonstrate this effect have been somewhat inconclusive due to the lack of quantitative agreement between theory and experiment. Figure 3 shows experimental and theoretical data for a

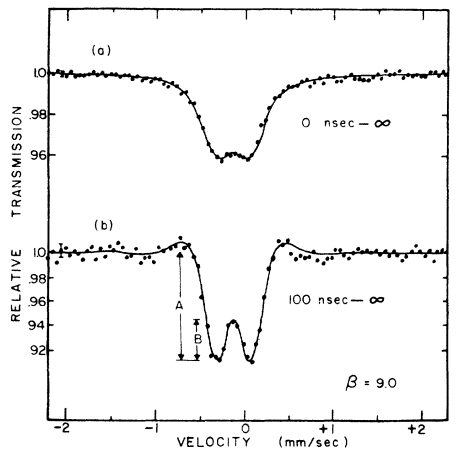

FIG. 3. Regular (a) and coincidence (b) Mössbauer spectra for Pb_2NbFeO_6.

Co^{57}-in-copper source and a Pb_2NbFeO_6 absorber. Figure 3(a) shows the regular Mössbauer results while Fig. 3(b) shows data obtained in delayed coincidence for those nuclei that decay after 100 nsec. The solid lines are computed spectra generated by superimposing two lines separated by 0.38 mm/sec and integrating the time-dependent intensity from 0 to ∞ and 100 nsec to ∞, respectively (numerically ∞ corresponds to 1000 nsec). The value of $\beta = 9.0$ was chosen to fit the observed regular Mössbauer linewidth. Notice that the valley-to-total absorption ratio (B/A) goes from $B/A \approx 0.05$ (regular Mössbauer) to $B/A \approx 0.40$ (coincidence-Mössbauer). In this example it is reasonably clear that two lines are present in the Mössbauer spectrum. If, however, a spectrum contains more lines, e.g., two sets of hyperfine patterns, the enhanced resolution demonstrated here may be essential for extracting the effective field parameters.

*Work supported in part by the U. S. Atomic Energy Commission under Contract No. AT (30-1)-3973, NYO-3973-1.

†Portions taken from a thesis by D.W.H. to be submitted in partial fulfillment of the Ph.D degree.

‡Summer research participant (1968) at the Oak Ridge National Laboratory.

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TESTS OF TIME-REVERSAL INVARIANCE OF STRONG INTERACTIONS UTILIZING THE MÖSSBAUER EFFECT*

J. P. Hannon and G. T. Trammell
Physics Department, Rice University, Houston, Texas
(Received 26 July 1968)

The currents induced in the inner electronic shells by a nuclear Mössbauer γ transition gives an $E(L+1)$–$M(L)$ phase difference on the order of 10^{-2}–10^{-3} rad which must be taken into account in the interpretation of experiments designed to detect such phase differences arising from possible failure of T invariance in nuclear interactions.

Recently two groups[1,2] have reported on experiments designed to detect the effect of T-noninvariant nuclear interactions by measuring the angular and polarization dependence of γ-ray absorption or emission using Mössbauer nuclei. Lloyd[3] first pointed out that in a nuclear transition in which a γ ray of mixed multipolarity (e.g., $M1$-$E2$) is emitted (or absorbed) the reduced matrix elements of the current multipole moments which give the amplitudes of the respective radiations (to lower order in e) are in phase (or 180° out of phase) if the strong (and electromagnetic) interactions are T invariant. On the other hand if there is a small T-noninvariant admixture in the interactions then the reduced matrix elements are no longer relatively real. For a mixed $M1$-$E2$ transition,

$$\langle f \| E2 \| i \rangle / \langle f \| M1 \| i \rangle = |\delta| \exp(i\eta), \tag{1}$$